

FORM PTO-1190 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER <b>146154.00056</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. <b>09/913373</b> New U.S. National Stage Patent Application
INTERNATIONAL APPLICATION NO. PCT/EP00/02875	INTERNATIONAL FILING DATE March 31, 2000	PRIORITY DATE CLAIMED April 20, 1999	
TITLE OF INVENTION BIAXIALLY ORIENTED FILM FOR THE PRODUCTION OF CERAMIC CAPACITORS			
APPLICANT(S) FOR DO/EO/US Karl-Heinz KOCHEM, Kerstin MÜLLER-NAGEL and Robert SCHMIDT			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</p> <p>4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).</p> <p>b. <input type="checkbox"/> has been communicated by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p>a. <input type="checkbox"/> is attached hereto.</p> <p>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</p> <p>b. <input type="checkbox"/> have been communicated by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p>			
Items 11 to 20 below concern document(s) or information included:			
<p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment.</p> <p>14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</p> <p>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</p> <p>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</p> <p>20. <input checked="" type="checkbox"/> Other items or information: PCT Request, Preliminary International Examination Report (in German) and PCT Notification Concerning Submission or Transmittal of Priority Document</p>			

U.S. APPLICATION NO. 09/913573

INTERNATIONAL APPLICATION NO.  
PCT/EP00/02875ATTORNEY'S DOCKET NUMBER  
146154.0005621. ☐ The following fees are submitted:**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):**Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
and International Search Report not prepared by the EPO or JPO. .... **\$100.00**International preliminary examination fee (37 CFR 1.482) not paid to  
USPTO but International Search Report prepared by the EPO or JPO. .... **\$860.00**International preliminary examination fee (37 CFR 1.482) not paid to USPTO  
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. .... **\$710.00**International preliminary examination fee (37 CFR 1.482) paid to USPTO  
but all claims did not satisfy provisions of PCT Article 33(1)-(4). .... **\$690.00**International preliminary examination fee (37 CFR 1.482) paid to USPTO  
and all claims satisfied provisions of PCT Article 33(1)-(4). .... **\$100.00****ENTER APPROPRIATE BASIC FEE AMOUNT =****CALCULATIONS PTO USE ONLY**

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than  
months from the earliest claimed priority date (37 CFR 1.492(e)). ☐ 20 ☐ 30

\$

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	8 - 20 =	0	x \$18.00	\$
Independent claims	2 - 3 =	0	x \$80.00	\$
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 860.00
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$
<b>SUBTOTAL =</b>				\$ 860.00
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$
<b>TOTAL NATIONAL FEE =</b>				\$ 860.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$
<b>TOTAL FEES ENCLOSED =</b>				\$ 860.00
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REGISTRATION NUMBER

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of Karl-Heinz KOCHER et al.	
New U.S. National Stage Patent Application of PCT/EP00/02875	Docket No.: 146154.00056
Filed: August 13, 2001	
For: BIAXIALLY ORIENTED FILM FOR THE PRODUCTION OF CERAMIC CAPACITORS	

**PRELIMINARY AMENDMENT**

Honorable Commissioner of Patents  
and Trademarks  
Washington, D.C. 20231

Sir:

Prior to examination of the above-identified application, Applicants respectfully  
request that the following amendments be entered into the application:

IN THE CLAIMS:

Please replace claims 3-7 with the following unmarked versions:

3. Use according to claim 1, characterized in that the film has a second top layer  
C, and the surface of this top layer C forms the smoother film surface.
4. Use according to claim 1, characterized in that the top layer C essentially  
consists of a propylene homopolymer and does not contain any antiblocking agent.

5. Use according to claim 1, characterized in that the top layer A comprises the propylene polymer in an amount of from 70 to 99.5% by weight and the incompatible polyolefin in an amount of from 0.5 to 30% by weight.
6. Use according to claim 1, characterized in that the film coated with a ceramic layer is provided with a metal layer on the surface of the ceramic layer.
7. Ceramic capacitor produced by means of a use according to claim 1.

### REMARKS

Applicants respectfully request that the foregoing amendments to claims 3-7 be entered in order to avoid this application incurring a surcharge for the presence of one or more multiple dependent claims.

Respectfully submitted,



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Registration No. 35,020  
Attorney for Applicant

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Washington, D.C. 20090-7223  
202-347-0066

Date: **Monday, August 13, 2001**

DWW/yea

Attachment:  
Appendix

## APPENDIX

**Version with Markings to Show Changes in the Claims**

Changes in the Claims:

The following is a marked-up version of the amended claims:

3. Use according to claim 1 ~~and/or 2~~, characterized in that the film has a second top layer C, and the surface of this top layer C forms the smoother film surface.
4. Use according to ~~one or more of claims 1 to 3~~ claim 1, characterized in that the top layer C essentially consists of a propylene homopolymer and does not contain any antiblocking agent.
5. Use according to ~~one or more of claims 1 to 4~~ claim 1, characterized in that the top layer A comprises the propylene polymer in an amount of from 70 to 99.5% by weight and the incompatible polyolefin in an amount of from 0.5 to 30% by weight.
6. Use according to ~~one or more of claims 1 to 5~~ claim 1, characterized in that the film coated with a ceramic layer is provided with a metal layer on the surface of the ceramic layer.
7. Ceramic capacitor produced by means of a use according to ~~one or more of claims 1 to 5~~ claim 1.

**Biaxially oriented film for the production of ceramic capacitors**

The invention relates to a biaxially oriented multilayer polyolefin film which is used as support film for ceramic layers. The invention furthermore relates to a process for the production of ceramic capacitors.

- 5 Ceramic capacitors are capacitors in which the dielectric consists of ceramic layers. In order to produce these capacitors, firstly a ceramic starting material, generally a ceramic powder, is converted into a highly viscous composition using suitable solvents and binders. In subsequent process steps, this composition is converted further into thin layers, which are subsequently provided with a metal layer (capacitor electrodes). These
- 10 ceramic layers covered in this way are formatted, stacked, pressed and finally fired at elevated temperatures in order to sinter the ceramic. The usable capacitor is produced from these blanks by subsequent provision of the electrodes with electrical contacts and encapsulation in a synthetic-resin coating (Spektrum der Wissenschaft September 1988, page 88 ff.).
- 15

- The ceramic layers may consist of different materials. Suitable ceramic materials are known in the prior art, such as, for example, metal oxides and/or titanates, for example barium titanate, magnesium silicate, titanium dioxide, bismuth oxide and mixtures thereof. These ceramics are distinguished by high dielectric constants.
- 20

- Various processes are known for producing the ceramic layers which are converted further into the capacitor. For example, the highly viscous, ceramic coating composition is firstly applied to a suitable flexible substrate and dried. This substrate has to be removed again in later processing
- 25 steps, during which the resultant ceramic layer or film must not be damaged during detachment. It is known in the prior art to use paper, polyester film or polypropylene film for this purpose.

- JP 06305041 A describes an oriented, multilayer film having a polypropylene base layer and ethylene-propylene copolymer top layers, whose surface roughness Ra is said to be in the range from 0.08 to 0.5  $\mu\text{m}$ . The roughness values of the two surfaces should differ by not more than 0.1  $\mu\text{m}$ . The ceramic layers generated on this film can only be dried at low temperatures, since otherwise the adhesion of the ceramic layer to the
- 30

film becomes too great and flaws are generated in the ceramic film on detachment of the support film.

5 JP 01196111 A describes a single-layer oriented polypropylene film which comprises polydimethylsiloxane as release agent with respect to the ceramic layer. However, the polydimethylsiloxane may be partly transferred onto the surface of the ceramic layer, where it impairs the adhesion of the metal coatings to this surface.

10 JP 60206620 H describes a film made from a mixture comprising polypropylene, polymethylpentene and HDPE. The film is said to have good release properties and surface roughness due to the HDPE.

The object of the present invention was to provide a film which is suitable as support film for ceramic layers. On the one hand, there should be adequate adhesion between the film and the ceramic layer in order that the ceramic material can be applied well and the coated film can be wound up.  
15 At the same time, it must be possible to detach the ceramic layer from the film after drying without damaging the ceramic layer, in particular its surface. Regarding the ceramic layer, it is required that this must have the smoothest possible surface after detachment from the film.

This object is achieved in accordance with the invention by the use of a  
20 polyolefinic, oriented multilayer film as support film in the production of a ceramic capacitor, where

- the multilayer film consists of a base layer and at least one top layer A, where the top layer A comprises a propylene polymer and at least one incompatible polyolefin, and the incompatible polyolefin is an LDPE, HDPE, MDPE, ethylene-propylene copolymer or [lacuna], and
  - 25 • the surface of the top layer A has greater roughness than the other surface of the film, and
  - the film is provided with a ceramic coating on the smoother surface of the film, and this coating is dried and subsequently separated from the support film.
- 30

The base layer of the multilayer film according to the invention essentially consists of a polyolefin, preferably of a propylene polymer, and optionally

added additives in effective amounts in each case. The base layer generally comprises at least 90% by weight, preferably from 95 to < 100% by weight, of the polyolefin.

- 5 The polypropylene polymer comprises predominantly (at least 90%) of propylene and has a melting point of 140°C or above, preferably from 150 to 170°C. Isotactic polypropylene having an n-heptane-soluble content of 6% by weight or less, copolymers of ethylene and propylene having an ethylene content of 5% by weight or less, and copolymers of propylene with C<sub>4</sub>-C<sub>8</sub>- $\alpha$ -olefins having an  $\alpha$ -olefin content of 5% by weight or less  
10 represent preferred propylene polymers for the base layer, particular preference being given to isotactic polypropylene. The propylene polymer of the base layer generally has a melt flow index of from 0.5 g/10 min to 15 g/10 min, preferably from 3 g/10 min to 8 g/10 min, at 230°C and a force of 21.6 N (DIN 53 735). The weight percentages indicated are based on the  
15 respective copolymer.

- The top layer A arranged on the surface of the base layer comprises a propylene polymer and at least one incompatible polyolefin. The incompatible polyolefin is generally present in the top layer in an amount of from 0.5 to 30% by weight, preferably from 1 to 25% by weight, in particular from 3 to  
20 15% by weight. The propylene polymer is present in an amount of from 70 to 99.5% by weight, preferably from 75 to 99% by weight, in particular from 85 to 97% by weight. The top layer may, if desired, additionally comprise additives in effective amounts in each case.

- The propylene polymer of the top layer A comprises predominantly (at least  
25 90%) propylene and has a melting point of 120°C or above, preferably from 140 to 170°C. Isotactic polypropylene having an n-heptane-soluble content of 6% by weight or less, copolymers of ethylene and propylene having an ethylene content of 10% by weight or less, and copolymers of propylene with C<sub>4</sub>-C<sub>8</sub>- $\alpha$ -olefins having an  $\alpha$ -olefin content of 5% by weight or less  
30 represent preferred propylene polymers for the base layer, particular preference being given to isotactic propylene homopolymer. The propylene polymer of the top layer generally has a melt flow index of from 0.5 g/10 min to 15 g/10 min, preferably from 3 g/10 min to 10 g/10 min, at 230°C and a force of 21.6 N (DIN 53 735). The weight percentages  
35 indicated are based on the respective copolymer. The MFI of the top-layer



polymer should be at least as great, in general greater (from about 25 to 100%), than the MFI of the base layer.

- 5 Incompatible polyolefins are those which are not completely miscible with the polypropylene of the top layer A and form a separate phase. This incompatibility causes a certain roughness of the surface, which is desired for the present invention. Suitable incompatible polyolefins are HDPE, MDPE, LDPE or syndiotactic polypropylenes or cycloolefin polymers.

- 10 For the invention, preference is given to high density polyethylenes (HDPEs) which have a melt flow index MFI, measured in accordance with ISO 1133 at 21.6 N/190°C, in the range from 0.1 to 2.0 g/10 min, preferably from 0.5 to 1.5 g/10 min, and a density, measured at 23°C in accordance with DIN 53 479, Method A, or ISO 1183, in the range from 0.935 to 0.97 g/cm<sup>3</sup>, preferably from 0.94 to 0.96 g/cm<sup>3</sup>, and a melting point, measured by DSC (maximum of the melting curve, heating rate 20°C/min), of  
15 between 120 and 150°C, preferably between 125 and 135°C.

- For the invention, preference is given to medium density polyethylenes (MDPEs) which have a melt flow index MFI, measured in accordance with ISO 1133 at 21.6 N/190°C, in the range from 0.1 to 3.0 g/10 min, preferably from 0.6 to 1.5 g/10 min, and a density, measured at 23°C in accordance  
20 with DIN 53 479, Method A, or ISO 1183, in the range from 0.925 to 0.94 g/cm<sup>3</sup>, preferably from 0.925 to 0.935 g/cm<sup>3</sup>, and a melting point, measured by DSC (maximum of the melting curve, heating rate 20°C/min), of between 115 and 145°C, preferably between 115 and 130°C.

- For the invention, preference is given to low density polyethylenes (LDPEs)  
25 which have a melt flow index MFI, measured in accordance with ISO 1133 at 21.6 N/190°C, in the range from 0.1 to 3.5 g/10 min, preferably from 0.5 to 2.0 g/10 min, and a density, measured at 23°C in accordance with DIN 53 479, Method A, or ISO 1183, in the range from 0.91 to 0.925 g/cm<sup>3</sup>, preferably from 0.915 to 0.925 g/cm<sup>3</sup>, and a melting point, measured by  
30 DSC (maximum of the melting curve, heating rate 20°C/min), of between 110 and 135°C, preferably between 110 and 125°C.

Cycloolefin polymers (COPs) are homopolymers built up from only one type of cycloolefin or copolymers built up from cycloolefins and

- comonomers (COCs), where the comonomer content is at most 50% by weight, based on the weight of the cycloolefin polymer. Cycloolefins are mono- or polyunsaturated polycyclic ring systems, such as cycloalkenes, bicycloalkenes, tricycloalkenes or tetracycloalkenes. The ring systems may be mono- or polysubstituted.

- Of the COPs described above, preference is given to those that are built up from monoalkylated or unsubstituted cycloolefins. Particularly preferred cycloolefin homopolymers are polynorbornene, polydimethyloctahydronaphthalene, polycyclopentene and poly(5-methyl)norbornene. The cycloolefin polymers may also be branched. Products of this type may have comb or star structures.

- If desired, the cycloolefins described above may also be copolymerized with comonomers. These cycloolefin copolymers (COCs) contain up to 50% by weight, preferably 1 – 35% by weight, in particular from 5 to 25% by weight, based on the weight of the COC, of comonomer. Preferred comonomers are olefins having from 2 to 6 carbon atoms, in particular ethylene and butylene.

- The cycloolefin polymers can be prepared with the aid of transition-metal catalysts. Preparation processes are described, for example, in DD-A-109 225, EP-A-0 407 870 and EP-A-0 485 893, which are expressly incorporated herein by way of reference.

- Syndiotactic polypropylenes are homopolymers or copolymers having a propylene content of at least 70% by weight, preferably greater than 80% by weight, in particular in the range from 95 to 100% by weight, based on the total weight of the polymer. The propylene content of the polymer has an isotacticity of < 15% by weight, in particular < 6%. The mean sequence length of the syndiotactic sequences are > 20%, preferably > 25%. Suitable comonomers of the copolymer are olefins having from 2 to 8 carbon atoms, of which ethylene and/or butylenes are preferred.

- The polypropylene and the incompatible polyolefin are employed as a mixture or as a blend. For the purposes of the present invention, mixtures are taken to mean mechanical mixtures prepared from the individual components. To this end, the individual constituents are generally poured

together as pressed moldings of small size, for example lenticular or spherical granules, and mixed mechanically using a suitable shaking device.

For the purposes of the present invention, a blend is an alloy-like compound of the individual components which can no longer be split into  
5 the original constituents. A blend has properties like a homogeneous material and can be correspondingly characterized by means of suitable parameters.

In accordance with the invention, the surface of the top layer A is rougher than the opposite film surface. It has been found that a rougher surface is  
10 essential for the processing behavior of the film in the use according to the invention. Due to the smooth opposite surface C, the film tends to block. In addition, the winding-up behavior is extremely problematic in the case of a smooth surface. In accordance with the prior art, antiblocking agents are incorporated into one of the top layers in order to overcome such problems.  
15 It has been found that incorporation of antiblocking agents into the top layer A is disadvantageous for the use according to the invention. It has been observed that embodiments comprising antiblocking agents in the top layer A lead to damaged ceramic layers, since the antiblocking agents in the top layer A leave impressions in the ceramic layer on winding-up of the coated  
20 film or may be transferred thereto. Ceramic layers damaged in this way can no longer be used for the production of capacitors.

In order to facilitate processing of the film without antiblocking agents and to avoid damage to the ceramic layer, a surface roughness of the top layer A is generated in accordance with the invention by means of a mixture of  
25 polypropylene and incompatible polyolefins. Surprisingly, the surface structure generated in this way is sufficiently rough in order to process the film well in spite of a smooth opposite surface. The film can be wound up and unwound with no problems without the incompatible polymer incorporated into the top-layer matrix being mechanically removed.

30 In contrast to the inorganic pigments usually employed for achieving a certain surface roughness, the structure of the rough surface of the film according to the invention does not result (in particular owing to the low hardness of the incompatible polymer) in any damage or surface deformations at all in the ceramic layer during winding up and unwinding. The use

according to the invention thus enables the production of thin ceramic layers which have a desirable smooth surface and have no flaws or other damage.

- 5 The thickness of the top layer A is greater than  $0.3\text{ }\mu\text{m}$  and is preferably in the range from  $0.4$  to  $3\text{ }\mu\text{m}$ , preferably from  $0.6$  to  $1.5\text{ }\mu\text{m}$ .

In accordance with the invention, the opposite surface of the film is smoother than the surface of the top layer A. This smooth surface may be the surface of the base layer. In a preferred embodiment, the film has a second top layer C by means of which the smoother surface is formed.

- 10 The top layer C essentially consists of a polyolefin, preferably a propylene polymer, and, if desired, added additives in effective amounts in each case. The top layer C generally comprises at least 90% by weight, preferably from 95 to < 100% by weight, of the polyolefin.

Examples of olefinic polymers of this type are

- 15 propylene homopolymer or  
a copolymer of  
ethylene and propylene or  
ethylene and 1-butylene or  
propylene and 1-butylene or  
20 a terpolymer of  
ethylene and propylene and 1-butylene or  
particular preference being given to  
random ethylene-propylene copolymers having  
an ethylene content of from 1 to 10% by weight, preferably from  
25 2.5 to 8% by weight, or  
random propylene-1-butylene copolymers having  
a butylene content of from 2 to 25% by weight, preferably from 4  
to 20% by weight,  
in each case based on the total weight of the copolymer, or  
30 random ethylene-propylene-1-butylene terpolymers having  
an ethylene content of from 1 to 10% by weight, preferably from  
2 to 6% by weight, and  
a 1-butylene content of from 2 to 20% by weight, preferably from  
4 to 20% by weight,

in each case based on the total weight of the terpolymer.

- 5 Suitable propylene homopolymers comprise predominantly (at least 90%) propylene and have a melting point of 140°C or above, preferably from 150 to 170°C, preference being given to isotactic homopolypropylene having an n-heptane-soluble content of 6% by weight or less, based on the isotactic homopolypropylene. The homopolymer generally has a melt flow index of from 0.5 g/10 min to 15 g/10 min, preferably from 1.5 g/10 min to 10 g/10 min, at 230°C and a force of 21.6 N (DIN 53 735).

- 10 The above-described copolymers and/or terpolymers employed in the top layer C generally have a melt flow index of from 1.5 to 30 g/10 min, preferably from 3 to 15 g/10 min. The melting point is in the range from 120 to 140°C. All the melt flow indices given above are measured at 230°C and a force of 21.6 N (DIN 53 735).

- 15 As part of the use according to the invention, the smoother surface of the top layer C is provided with a ceramic coating. It has been found that the polyolefinic surface has adequate adhesion in order for a thin layer of ceramic material to be applied thereto. At the same time, however, the adhesion between the surface of the film and the ceramic layer is sufficiently low, after corresponding drying and/or processing steps, for the support film to be separated from the ceramic layer without damaging the ceramic layer produced in this way. Embodiments having a homopolymer top layer have proven particularly advantageous since these have greater heat resistance than copolymer or terpolymer layers and thereby enable higher drying and/or processing temperatures in the production of the ceramic layer. In addition, it has been found that the ceramic layer can be separated from the support layer having a homopolymer top layer with fewer problems.

- 30 The top layer C may comprise conventional neutralizers, stabilizers, if desired antistatics and lubricants. It is essential for the use according to the invention that the top layer C does not comprise any antiblocking agents that increase the surface roughness. The increased surface roughness would be transferred to the surface of the ceramic layer during coating. However, the ceramic layers should have the smoothest possible surface for use as a dielectric in order to avoid flaws and air bubbles. In addition, it

has been found that the antiblocking agent particles remain stuck in the ceramic layer during detachment of the support film! This is extremely undesired since the antiblocking particles in the ceramic layer result in electrical flaws and air bubbles.

- 5 The thickness of the top layer C is greater than 0.3  $\mu\text{m}$  and is preferably in the range from 0.4 to 3  $\mu\text{m}$ , preferably from 0.6 to 1.5  $\mu\text{m}$ .

- The total thickness of the multilayer polyolefin film according to the invention can vary within broad limits and depends on the intended use. It is from 6 to 70  $\mu\text{m}$ , preferably from 10 to 50  $\mu\text{m}$ , with the base layer making  
10 up from about 50 to 98% of the total film thickness.

- In order to improve certain properties of the polyolefin film still further, both the base layer and the top layer(s) may comprise further additives in an effective amount in each case, preferably antistatics and/or lubricants and/or stabilizers and/or neutralizers. All amount data in the following list are in per cent by weight (% by weight), in each case based on the layer or  
15 layers to which the additive can be added.

- Preferred antistatics are alkali metal alkanesulfonates, polyether-modified, i.e., ethoxylated and/or propoxylated polydiorganosiloxanes (polydialkylsiloxanes, polyalkylphenylsiloxanes and the like) and/or essentially straight-chain and saturated aliphatic, tertiary amines containing an aliphatic radical  
20 having from 10 to 20 carbon atoms which are substituted by  $\square$ -hydroxy-(C<sub>1</sub>-C<sub>4</sub>)alkyl groups, where N,N-bis(2-hydroxyethyl)alkylamines having from 10 to 20 carbon atoms, preferably from 12 to 18 carbon atoms, in the alkyl radical are particularly suitable. The effective amount of antistatic is in the  
25 range from about 0.05 to 0.3% by weight.

- Lubricants are higher aliphatic acid amides, higher aliphatic acid esters, waxes and metal soaps, as well as polydimethylsiloxanes. The effective amount of lubricant is in the range from 0.1 to 3% by weight. Particularly  
30 suitable is the addition of higher aliphatic acid amides in the range from 0.15 to 0.25% by weight in the base layer and/or top layers. A particularly suitable aliphatic acid amide is erucamide. The addition of polydimethylsiloxanes is preferred in the range from 0.3 to 2.0% by weight, in particular polydimethylsiloxanes having a viscosity from 10,000 to 1,000,000 mm<sup>2</sup>/s.

- Stabilizers which can be employed are the conventional compounds having a stabilizing action for polymers of ethylene, propylene and other  $\alpha$ -olefins. Their added amount is between 0.05 and 2% by weight. Particularly suitable are phenolic stabilizers, alkali metal or alkaline earth metal stearates and/or alkali metal or alkaline earth metal carbonates. Phenolic stabilizers are preferred in an amount from 0.1 to 0.6% by weight, in particular from 0.15 to 0.3% by weight, and having a molecular weight of greater than about 500 g/mol. Pentaerythrityl tetrakis-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate or 1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)benzene are particularly advantageous. Also preferred are alkaline earth metal stearates and/or carbonates in an added amount of from 0.01 to 0.05% by weight, in particular calcium stearate and/or calcium carbonate having a mean particle size of less than 0.1 mm, preferably from 0.03 to 0.07 mm, an absolute particle size of less than 5  $\mu$ m and a specific surface area of at least 40 m<sup>2</sup>/g.

- As part of the investigations regarding the present invention, it has been found that the film can also be employed in the application of photoresists. Photoresist layers are usually applied to a suitable film material and wound up with this support. In some cases, the adhesion between the support film and the applied photoresist layer is too great, and problems may consequently arise during unwinding and further processing. It has been found that the film described above can advantageously be inserted into the reel as separating film in order to prevent adhesion between the support film and the photoresist layer.

- In order to produce the multilayer film by the coextrusion process, which is known per se, the melts corresponding to the individual layers of the film are coextruded through a flat-film die, the resultant film is taken off over one or more roll(s) for solidification, the film is subsequently biaxially stretched (oriented), the biaxially stretched (oriented) film is heat-set and wound up.

- The biaxial stretching (orientation) can be carried out simultaneously or sequentially, with sequential biaxial stretching, in which stretching is firstly carried out longitudinally (in the machine direction) and then transversely (perpendicular to the machine direction), being preferred.

Firstly, as is conventional in the coextrusion process, the polymer or polymer mixture or the blend of the individual layers is compressed and liquefied in an extruder, it being possible that any additives have already  
5 been added to the polymer. The melts are then forced simultaneously through a flat-film die (slot die), and the extruded multilayer film is taken off on one or more take-off rolls, during which it cools and solidifies.

The film obtained in this way is then stretched longitudinally and  
10 transversely to the extrusion direction, which results in alignment of the molecule chains. The stretching in the longitudinal direction is preferably carried out at a ratio of from 4:1 to 7:1 and the stretching in the transverse direction is preferably carried out at a ratio of from 8:1 to 10:1. The longitudinal stretching is advantageously carried out with the aid of two rolls  
15 running at different speeds corresponding to the target stretching ratio, and the transverse stretching is advantageously carried out with the aid of an appropriate tenter frame.

The biaxial stretching of the film is followed by heat-setting (heat treatment)  
20 thereof, in which the film is held at a temperature from 150 to 160°C for from about 0.5 to 10 seconds. The film is subsequently wound up in a conventional manner by means of a wind-up unit.

It has proven particularly favorable to keep the take-off roll or rolls by  
25 means of which the extruded film is cooled and solidified at an elevated temperature compared to the prior art of from 10 to 60°C by means of a heating and cooling circuit.

In addition, the biaxial stretching is also advantageously carried out at an elevated film temperature, the longitudinal stretching preferably at from 90  
30 to 140°C and the transverse stretching preferably at from 150 to 190°C.

The invention furthermore relates to a process for the production of ceramic capacitors in which the film described above is used in accordance with the invention. For this purpose, firstly a ceramic starting material, generally a ceramic powder, is converted into a highly viscous composition  
35 using suitable solvents and binders. Suitable ceramic materials are, for example, metal oxides and/or titanates, for example barium titanate,



magnesium silicate, titanium dioxide, bismuth oxide and mixtures thereof. These ceramics are distinguished by high dielectric constants. The highly viscous, ceramic coating composition is applied to the smooth surface C of the film and dried. The film coated in this way is subsequently wound up.

- 5 The ceramic surface can, if desired, be metallized in a further processing step. The film is finally separated from the ceramic layer. The ceramic layers produced in this way are then converted further into ceramic capacitors in a manner known per se.

- 10 The invention is now explained in greater detail with reference to working examples.

#### Example 1

A three-layer film having an ABC layer structure is extruded by the coextrusion method from a slot die at an extrusion temperature of 250°C.

- 15 The base layer essentially consists of a propylene homopolymer having an n-heptane-soluble content of 4.5% by weight and a melting point of 163°C. The melt flow index of the propylene homopolymer is 3.3 g/10 min at 230°C and a load of 21.6 N (DIN 53 735, ISO 1133). The base layer comprises 0.13% by weight of pentaerythrityl tetrakis-4-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate (®Irganox 1010) for stabilization and 0.06% by
- 20 weight of calcium stearate as neutralizer. The base layer furthermore comprises 0.14% by weight of N,N-bis(2-hydroxyethyl)(C<sub>10</sub>-C<sub>20</sub>)alkylamine (®Armostat 300) as antistatic and an erucamide (Armoslip) in an amount of 0.2% by weight as lubricant.

- 25 The top layers A essentially consist of a mixture of 93% by weight of propylene homopolymer and 7% by weight of LDPE. The propylene homopolymer had an n-heptane-soluble content of 5% by weight and a melting point of 164°C and a melt flow index of 8.0 g/10 min at 230°C and a load of 21.6 N (DIN 53 735, ISO 1133). The propylene homopolymer comprised 0.05% by weight of Irgafos 168 (tris(2,4-di-tert-butyl phenyl phosphite) and 0.07% by weight of Irganox 1010 for stabilization and
- 30 0.06% by weight of calcium stearate as neutralizer. The LDPE had a melting point of 113°C (DSC) and a melt flow index of 0.85 g/10 min at 190°C and a load of 21.6 N (DIN 53 735, ISO 1133) and a density of 0.923 g/cm<sup>3</sup>.

The top layer C essentially consists (i.e. about 100% by weight) of a propylene homopolymer. The propylene homopolymer had an n-heptane-soluble content of 5% by weight and a melting point of 164°C and a melt flow index of 8.0 g/10 min at 230°C and a load of 21.6 N (DIN 53 735, ISO 1133). The propylene homopolymer comprised 0.05% by weight of Irgafos 168 (tris(2,4-di-tert-butyl phenyl phosphite) and 0.07% by weight of Irganox 1010 for stabilization and 0.06% by weight of calcium stearate as neutralizer.

After coextrusion, the extruded three-layer film is taken off over a first take-off roll and a subsequent take-up roll and cooled, subsequently pre-warmed, stretched longitudinally, stretched transversely, set and wound up over the corresponding process steps, with the following conditions being selected:

- |    |                          |  |
|----|--------------------------|--|
| 15 | Extrusion:               | Extrusion temperature 250°C<br>Temperature of the first take-off roll 35°C<br>Temperature of the take-up roll 29°C |
|    | Longitudinal stretching: | Preheating zone T = 145°C<br>Stretching roll T = 145°C<br>Longitudinal stretching by the factor 4.6                |
| 20 | Transverse stretching:   | Heating fields T = 182°C<br>Stretching fields T = 162°C<br>Transverse stretching by the factor 9.2                 |
|    | Setting:                 | Temperature T = 120°C  |

The film has a thickness of about 45 µm, with the base layer having a thickness of 43 µm and each top layer having a thickness of 1.0 µm.

#### Comparative Example 1

Example 1 is repeated. Compared with Example 1, only the composition of the top layer A was changed. The top layer A essentially consisted of the top-layer homopolymer described in Example 1. In addition, the top layer comprised 0.38% by weight of an antiblocking agent comprising SiO<sub>2</sub> having a mean particle diameter of 4.8 µm (Sylobloc 45). The process conditions were essentially unchanged.

- The film according to the example according to the invention is highly suitable as support film for ceramic coating. The film can be coated well. The ceramic layer can be detached very well after drying and has a smooth surface without flaws. Electron photomicrographs show well that the
- 5 the surface of the top layer A has uniform roughness, whereas the surface A of the film according to the comparative example exhibits scattered particles (antiblocking agent) which project out of the top layer. The elevations press through in an undesired manner onto the opposite side and into the surface of the ceramic coating. Disadvantageous flaws form in the ceramic layer in
- 10 this case.

The following table shows the other properties of the polyolefin films of the example and comparative example.

Table 1

Film property	Unit	Example	Comparative Example
Modulus of elasticity, longitudinal	N/mm <sup>2</sup>	2186	2349
Modulus of elasticity, transverse	N/mm <sup>2</sup>	4701	4659
Tear strength, longitudinal	N/mm <sup>2</sup>	148	131
Tear strength, transverse	N/mm <sup>2</sup>	358	306
Elongation at break, longitudinal	%	262	253
Elongation at break, transverse	%	54	45
Roughness Rz of the surface of top layer A	μm	0.56	0.72
Roughness Rz of the surface of top layer C	μm	0.3	0.33
Roughness Rmax of the surface of top layer A	μm	0.64	1.41
Roughness Rmax of the surface of top layer C	μm	0.37	0.41
Sliding friction A against C		0.33	0.45

15

The following measurement methods were used to characterize the raw

materials and the films:

Melt flow index

DIN 53 735 at a load of 21.6 N and 230°C.

5

Melting point

DSC measurement, maximum of the melting curve, heating rate 20°C/min.

Roughness

- 10 The roughness was determined in accordance with DIN 4768 as Rz and Rmax value with a cut-off of 2.5 mm.

Friction

The sliding friction was determined in accordance with DIN 53 375 at 23°C.

15

Modulus of elasticity

The modulus of elasticity is determined at the earliest 10 days after production in accordance with EN ISO 521-1 on a sample having a size of 15\*100 mm<sup>2</sup>.

20

Tear strength, elongation at break

The tear strength and elongation at break are determined in accordance with EN ISO 521-1 on a sample size of 15\*100 mm<sup>2</sup>.

25 Shrinkage:

- The longitudinal and transverse shrinkage values relate to the respective length elongation of the film (longitudinally L<sub>0</sub> and transversely T<sub>0</sub>) before the shrinkage process. The longitudinal direction is the machine direction, and the transverse direction is defined correspondingly as the direction transverse to the machine direction. The 10\*10 cm<sup>2</sup> specimen is shrunk in a fan-assisted oven at the respective temperature (from 100 to 140°C) over a period of 15 minutes. The remaining length elongations of the specimen are subsequently re-determined longitudinally and transversely (L<sub>1</sub> and T<sub>1</sub>). The shrinkage in % is then quoted as the difference in the measured length elongations divided by the original length L<sub>0</sub> and T<sub>0</sub> times 100.
- 30
- 35

$$\text{Longitudinal shrinkage } L_s [\%] = \frac{L_0 - L_1}{L_0} * 100 [\%]$$

$$\text{Transverse shrinkage } T_s [\%] = \frac{T_o - T_l}{T_o} * 100 [\%]$$

- 5 This determination method for the longitudinal and transverse shrinkage corresponds to DIN 40634.

#### Density

The density is determined in accordance with DIN 53479, Method A.

## Patent Claims

1. Use of a polyolefinic, oriented multilayer film as support film in the production of a ceramic capacitor, characterized in that

- 5                   • the multilayer film consists of a base layer and at least one top layer A, where the top layer A comprises a propylene polymer and at least one incompatible polyolefin, and in that the incompatible polyolefin is an LDPE, HDPE, MDPE, ethylene-propylene copolymer or a cycloolefin polymer or a syndiotactic polymer, and
- 10                  • the surface of the top layer A has greater roughness than the opposite surface of the film, and
- the film is provided with a ceramic coating on the smoother surface of the film, and this coating is dried and subsequently separated from the support film.

- 15           2. Use according to claim 1, characterized in that the top layer A does not contain any particulate antiblocking agent.

3. Use according to claim 1 and/or 2, characterized in that the film has a second top layer C, and the surface of this top layer C forms the smoother film surface.

- 20           4. Use according to one or more of claims 1 to 3, characterized in that the top layer C essentially consists of a propylene homopolymer and does not contain any antiblocking agent.

5. Use according to one or more of claims 1 to 4, characterized in that the top layer A comprises the propylene polymer in an amount of from 70 to 99.5% by weight and the incompatible polyolefin in an amount of from 0.5 to 30% by weight.
- 25

6. Use according to one or more of claims 1 to 5, characterized in that the film coated with a ceramic layer is provided with a metal layer on the surface of the ceramic layer.

7. Ceramic capacitor produced by means of a use according to one or more of claims 1 to 5.

8. Use of a polyolefinic, oriented multilayer film as support film in the production of photoresist layers, characterized in that

- 5
- the multilayer film consists of a base layer and at least one top layer A, where the top layer A comprises a propylene polymer and at least one incompatible polyolefin, and in that the incompatible polyolefin is an LDPE, HDPE, MDPE, ethylene-propylene copolymer or a cycloolefin polymer or a syndiotactic polymer, and
- 10
- the surface of the top layer A has greater roughness than the opposite surface of the film, and
  - the film, as separating film, is brought into contact with the photoresist layer with its smoother surface.



Docket No.  
146154.00056

# Declaration and Power of Attorney For Patent Application

## English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

**BIAXIALLY ORIENTED FILM FOR THE PRODUCTION OF CERAMIC CAPACITORS**

the specification of which

(check one)

☐ is attached hereto.

☒ was filed on March 31, 2000 as United States Application No. or PCT International

Application Number PCT/EP00/02875

and was amended on \_\_\_\_\_

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

19917790.2

GERMANY

20/04/1999

☐

(Number)

(Country)

(Day/Month/Year Filed)

☐

(Number)

(Country)

(Day/Month/Year Filed)

☐

(Number)

(Country)

(Day/Month/Year Filed)



I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or-PCT International filing date of this application:

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Status)  
(patented, pending, abandoned)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Status)  
(patented, pending, abandoned)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Status)  
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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